Review #1

We wish to thank the referee for his/her helpful comments. His/her full comments are copied hereafter in normal black font, and our responses are inserted in between in blue and bold font.

Overall, it is clear that a large amount of work was done by the authors. However, in its current state, I cannot recommend publication. I do encourage the authors to carefully consider my comments and resubmit the revised manuscript.

A major concern of mine is that this article doesn't present new science, so it may be better suited to another journal, such as Earth System Science Data (ESSD). The title and introduction actually sound somewhat appropriate for ACP, but the manuscript content is really not appropriate for ACP.

We feel that this comment, and also some of the comments below, are driven by the fact that the title of the manuscript was misleading. The main part of the analysis assess the potential of the satellite data to support the estimate of the decrease of the NO_x emissions over time, rather than to the analysis of this decrease itself. The title has been revised accordingly, and the introduction and conclusion have also been improved to make it clearer.

Another issue is that the manuscript is poorly organized and, therefore, difficult to read. ACP gives handy recommendations on the structure of a manuscript: https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html. I highly suggest that the authors review these recommendations. Here are two examples of what I mean:

- The title doesn't reflect what the paper is about. The paper title indicates that the paper is focused on discussing the causes of trends in European NOx emissions. At least, that's what I thought when reading the title. This isn't the case. Please revise the title to reflect the actual manuscript content.

We agree that the title of the article was potentially misleading and we have changed it. It is now: "Assessing the ability to quantify the decrease of NO_x anthropogenic emissions in 2019 compared to 2005 using OMI and TROPOMI satellite observations".

- The paper is wordy (by 30-50%); the introduction is particularly wordy and needs paragraph indents for clarity. Note that a goal of technical writing is to clearly and concisely convey a particular message. As an example, the purpose of your paper should be given in one sentence, such as in the abstract and as a topic sentence of a paragraph in the introduction. I had to piece it together over a very long paragraph, but I still don't know the overall goal of the paper given the vague title, vague abstract, and vague conclusions. An overview diagram of the steps in your work would be helpful.

We had made strong efforts to build an introduction providing a comprehensive overview of the context for the study: the NO_2 concentration and NO_x emission monitoring, the reduction of NO_x emissions, the state-of-the-art methods for the quantification of the NO_x emissions and the main limitations of both bottom-up inventory and of atmospheric inversions using satellite observations. We nevertheless understand the reviewer's comments and we have shortened and reformulated the introduction. The technical paragraph about

the similarities and the differences between the OMI and the TROPOMI algorithms and datasets has been moved to Section 2.

The objectives of the study have been reformulated and a diagram providing an overview of the inversion and experimental frameworks, and of the results has been inserted as the new Figure 1. We have also indented paragraphs in the introduction for clarity.

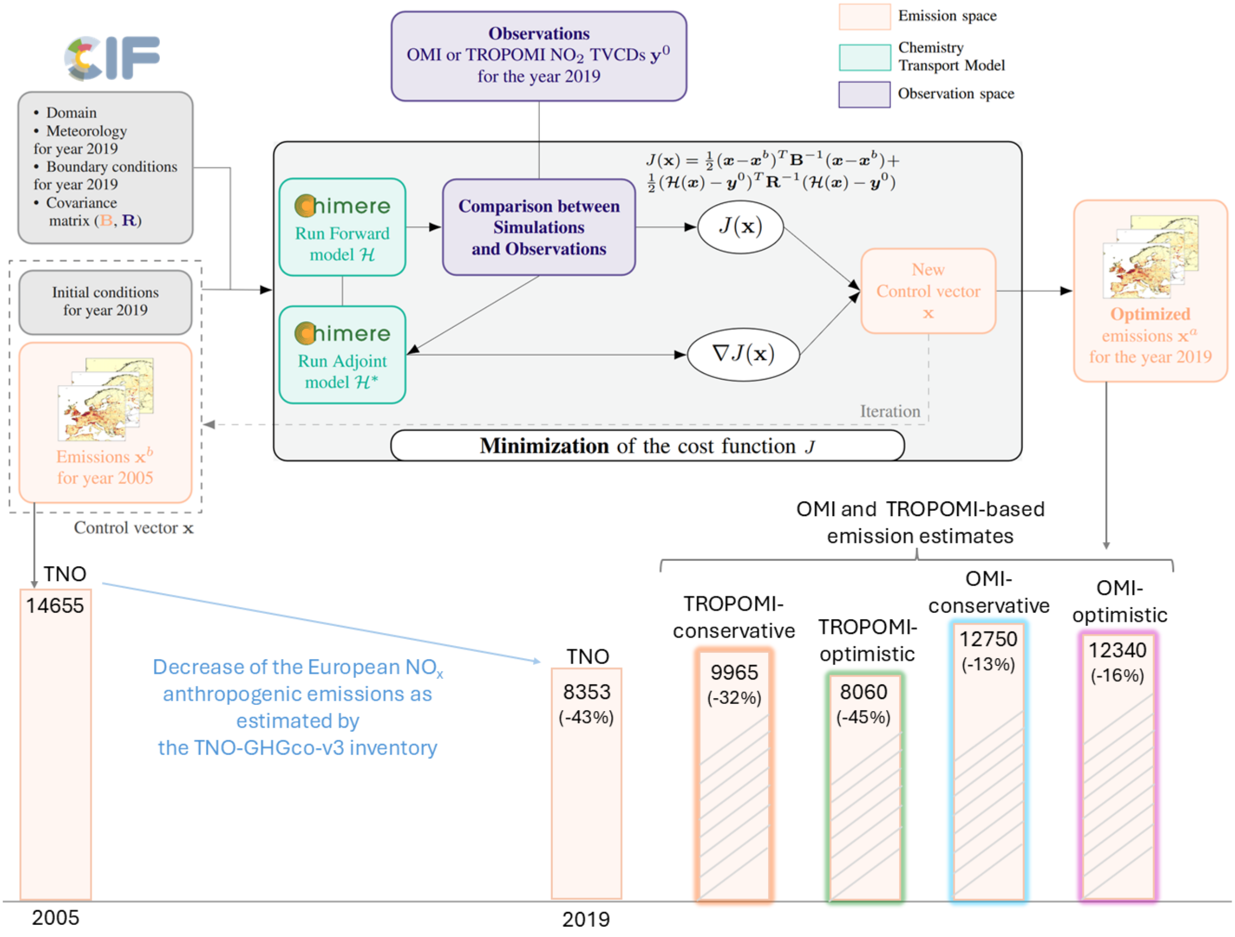

Figure 1. Simplified scheme of the iterative minimization in the CIF-CHIMERE inversion system and illustration of the decrease of the European NO_x emissions estimated both by TNO and by the OMI and TROPOMI-based inversions. The different inversions are described in Table 1. The numbers express the anthropogenic NO_x emission estimates for the EU-27+UK area in kteqNO_2. The numbers in brackets express the difference of the anthropogenic NOx emission estimates for the EU-27+UK in 2019 compared to 2005, in %.

Another major concern is the lack of validation of the data products and emission estimates. Why don't you compare results with independent in situ observations, such as from the Pandora network? That would certainly strengthen your conclusions about how your method impacts the emissions estimates. In fact, you say in the last sentence of the abstract "…our results —with OMI and TROPOMI data leading to different magnitudes of corrections on NOx anthropogenic emissions—suggest that more observational constraints would be required to sharpen the European emission estimates." You haven't even used the existing observational constraints !

The comparison between our type of regional CTM (here at 0.5° resolution) and typical in situ air quality observations in view to evaluate the emission estimates, and consequently the

validation of our inversions using such data or the assimilation of such data can be complex due, e.g., to the difficult representation of these observation close to the surface and to source in the model. As described by Lamsal et al. (2008), the instrument most commonly used for routine measurements of NO_2 is a chemiluminescence analyzer, a measurement technique which exhibits significant interference from other reactive oxidized nitrogen-containing species (NO_z) such as peroxyacetyl nitrate (PAN) and nitric acid (HNO_3). It could consequently result in an overestimation of ambient NO_2 concentrations (Savas et al. 2023). The joint assimilation of surface and satellite observation would raise further theoretical and technical challenges. Specific studies are needed to properly handle such a use of the in situ observations, which explains why we have a long list of publications focusing on the use of the satellite data (see our introduction, van der A et al., 2008; Lamsal et al., 2011; Mijling et al., 2012; Mijling et al., 2013; Miyazaki et al., 2017; Plauchu et al., 2024; van der A et al., 2024).

Furthermore, here, the analysis are focused on the assessment of the ability to provide information on the decrease of NO_x anthropogenic emissions in 2019 compared to 2005 based on satellite NO_2 observation (that from the OMI and TROPOMI instruments). Given their spatial coverage and spatial resolution, and their vertical representativity, these satellite observations— are likely more suitable for inversions at the targeted scales than the surface measurements in the area of analysis. This rationale is now better expressed with our new title and in our revision of the introduction.

Lamsal, L.N.; Martin, R.V.; van Donkelaar, A.; Steinbacher, M.; Celarier, E.A.; Bucsela, E.; Dunlea, E.J.; Pinto, J.P. Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument. J. Geophys. Res. 2008, 113, D16308.

Section 2.1: What are the strengths and limitations of this inversion system for your work? Has it been applied and validated with independent observations (e.g., Pandora)? Why are you using it relative to other inversion systems?

As now stated in the introduction, our inversion system combines both the advantages:

- **of solving large dimensional inversion problems, i.e. controlling emissions at relatively high temporal and spatial resolution and assimilating a large amount of observations, via the variational mode of the CIF**
- **of simulating NO_2 concentrations and the sensitivities of NO_2 tropospheric columns to surface emissions at a relatively high spatial resolution with a chemistry scheme, based on the Eulerian regional CTM CHIMERE, and its adjoint code.**

Our inversion system is therefore well designed to estimate NO_x emissions at the 0.5° spatial resolution and assimilate satellite NO_2 retrievals, taking into account the non-linearities of the NO_x chemistry. The CIF-CHIMERE variational inversion configuration has already been used for the estimation of the emissions of NO_x (Savas et al., 2023; Plauchu et al., 2024) but also of other species such as CO (Fortems-Cheiney et al., 2023) and CO_2 (MacGrath et al., 2023). These information have been added in the introduction.

Our study assesses the potential of the assimilation of OMI and TROPOMI satellite observations to provide information on the decrease of NO_x anthropogenic emissions in 2019

compared to 2005: we evaluate our emissions estimates for the year 2019 through comparisons to the independent estimates from the TNO inventory for this same year, at the European and at the national scales.

The title of Section 3.4 has been changed and is now: "Posterior estimates of NOx European anthropogenic emissions in 2019: evaluation with comparisons to the TNO-GHGco-v3 inventory"

Fortems-Cheiney, A., Broquet, G., Potier, E., Plauchu, R., Berchet, A., Pison, I., Denier van der Gon, H., and Dellaert, S.: CO anthropogenic emissions in Europe from 2011 to 2021: insights from Measurement of Pollution in the Troposphere (MOPITT) satellite data, Atmos. Chem. Phys., 24, 4635–4649, https://doi.org/10.5194/acp-24-4635-2024, 2024.

Plauchu, R., Fortems-Cheiney, A., Broquet, G., Pison, I., Berchet, A., Potier, E., Dufour, G., Coman, A., Savas, D., Siour, G., and Eskes, H.: NOx emissions in France in 2019–2021 as estimated by the high spatial resolution assimilation of TROPOMI NO2 observations, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2024-103, 2024.

McGrath, M. J., Petrescu, A. M. R., Peylin, P., Andrew, R. M., Matthews, B., Dentener, F., Balkovič, J., Bastrikov, V., Becker, M., Broquet, G., Ciais, P., Fortems-Cheiney, A., Ganzenmüller, R., Grassi, G., Harris, I., Jones, M., Knauer, J., Kuhnert, M., Monteil, G., Munassar, S., Palmer, P. I., Peters, G. P., Qiu, C., Schelhaas, M.-J., Tarasova, O., Vizzarri, M., Winkler, K., Balsamo, G., Berchet, A., Briggs, P., Brockmann, P., Chevallier, F., Conchedda, G., Crippa, M., Dellaert, S. N. C., Denier van der Gon, H. A. C., Filipek, S., Friedlingstein, P., Fuchs, R., Gauss, M., Gerbig, C., Guizzardi, D., Günther, D., Houghton, R. A., Janssens-Maenhout, G., Lauerwald, R., Lerink, B., Luijkx, I. T., Moulas, G., Muntean, M., Nabuurs, G.-J., Paquirissamy, A., Perugini, L., Peters, W., Pilli, R., Pongratz, J., Regnier, P., Scholze, M., Serengil, Y., Smith, P., Solazzo, E., Thompson, R. L., Tubiello, F. N., Vesala, T., and Walther, S.: The consolidated European synthesis of CO2 emissions and removals for the European Union and United Kingdom: 1990–2020, Earth Syst. Sci. Data, 15, 4295–4370, https://doi.org/10.5194/essd-15-4295-2023, 2023.

Savas, D., Dufour, G., Coman, A., Siour, G., Fortems-Cheiney, A., Broquet, G., Pison, I., Berchet, A., & Bessagnet, B. (2023). Anthropogenic NOx Emission Estimations over East China for 2015 and 2019 Using OMI Satellite Observations and the New Inverse Modeling System CIF-CHIMERE. Atmosphere, 14(1), 154. https://doi.org/10.3390/atmos14010154